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New Sensors for In-Pile Temperature Detection at the Advanced Test Reactor National Scientific User Facility

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ABSTRACT

The U.S. Department of Energy (DOE) designated the Advanced Test Reactor (ATR) as a National Scientific User Facility (NSUF) in April 2007 to support U.S. research in nuclear science and technology. As a user facility, the ATR is supporting new users from universities, laboratories, and industry, as they conduct basic and applied nuclear research and development to advance the nation's energy security needs. A key component of the ATR NSUF effort is to develop and evaluate new in-pile instrumentation techniques that are capable of providing measurements of key parameters during irradiation. This paper describes the strategy for determining what instrumentation is needed and the program for developing new or enhanced sensors that can address these needs. Accomplishments from this program are illustrated by describing new sensors now available and under development for in-pile detection of temperature at various irradiation locations in the ATR.

KEYWORDS

In-pile Sensors, High Temperature Irradiation Resistant Instrumentation

1. INTRODUCTION

The U.S. Department of Energy (DOE) designated the Advanced Test Reactor (ATR) as a National Scientific User Facility (NSUF) in April 2007 to support U.S. research in nuclear science and technology. By supporting users from universities, laboratories, and industry, the ATR will support basic and applied nuclear research and development and advance the nation's energy security needs. A key component of the ATR NSUF effort is to develop and implement in-pile instrumentation that is capable of providing measurements of key parameters during irradiation. This paper describes the strategy for identifying instrumentation needed for ATR irradiations and the program initiated to develop and evaluate new temperature sensors as part of this effort.

1.1. ATR Design and Irradiation Capabilities

The ATR is a versatile tool for conducting nuclear reactor, nuclear physics, reactor fuel, and structural material irradiation experiments. [1]

The ATR's maximum power rating is 250 MW_{th} with a maximum unperturbed thermal neutron flux of 1 $x10^{15}$ n/cm^2-s and a maximum fast neutron flux of 5 x 10^{14} n/cm^2-s. Because most contemporary experimental objectives do not require the upper limits of its capability, the ATR typically operates at lower power levels (nominally 110 MW_{th}). The ATR is available over 70% of the year, in cycles that typically range from 6 to 8 weeks, with outages lasting one or two weeks. The ATR is cooled by pressurized (2.5 MPa/360 psig) water that enters the reactor vessel bottom at an average temperature of 52 °C (126 °F), flows up outside cylindrical tanks that support and contain the core, passes through concentric thermal shields into the open part of the vessel, and then flows down through the core to a flow distribution tank below the core. When the reactor is operating at full power, the primary coolant has a maximum flowrate of 3.09 m^3/sec (49,000 gpm) and exits the vessel at 71 °C (160 °F).

As shown in Figure 1, the ATR core consists of 40 curved plate fuel elements in a serpentine arrangement around a 3 x 3 array of nine large high-intensity neutron flux traps. The unique ATR control device design permits large power variations among its nine flux traps using a combination of control cylinders (drums) and neck shim rods. The beryllium control cylinders contain hafnium plates that can be rotated toward and away from the core. Hafnium shim rods, which withdraw vertically, are inserted or withdrawn for minor power adjustments. Within bounds, the power level in each corner lobe of the reactor can be controlled independently to allow for different power and flux levels in the four corner lobes during the same operating cycle. The ratio of fast to thermal flux can be varied from 0.1 to 1.0. In addition to the nine large volume (up to 121.9 cm /48.0 in. long and up to 12.7 cm/5.0 in. diameter) high-intensity neutron flux traps, there are 66 irradiation positions inside the reactor core reflector tank, and two capsule irradiation tanks outside the core with 34 low-flux irradiation positions.

There are three basic types of test assembly configurations used in the ATR:

- ***Static Capsule Experiment*** – These capsules may contain a number of small sample or engineered components. Static capsule experiments may be sealed or may be in open configuration and contain material that can be in contact with the ATR primary coolant. Capsules may be any length, up to 121.9 cm (48.0 in.) and may be irradiated in any core position, including the flux traps. Irradiation temperature may be influenced by providing a gas gap in the capsule with a known thermal conductance. Peak temperatures may be measured using a series of melt wires, temperature-sensitive paint spots, or silicon carbide temperature monitors. Accumulated neutron fluences may be verified using flux wires.

- ***Instrumented Lead Experiment*** - Active control of experiments and data from test capsules during irradiation is achieved using core positions with instrumentation cables and temperature control gases in ATR instrumented lead experiments. Such experiments can have instrumentation, such as thermocouples, connected to individual capsules or single specimens. This instrumentation can be used to control and sample conditions within the capsule. For example, temperature control in individual zones is performed by varying the gas mixture (typically helium and neon) in the gas gap that thermally links the capsule to the water-cooled reactor structure. In addition to temperature, instrumented leads have been used to monitor the gas around the test specimen. In a fueled experiment, the presence of fission gases due to fuel failures or oxidation can be detected via gas chromatography. Instrument leads also allow real time display of experimental parameters in the control room.

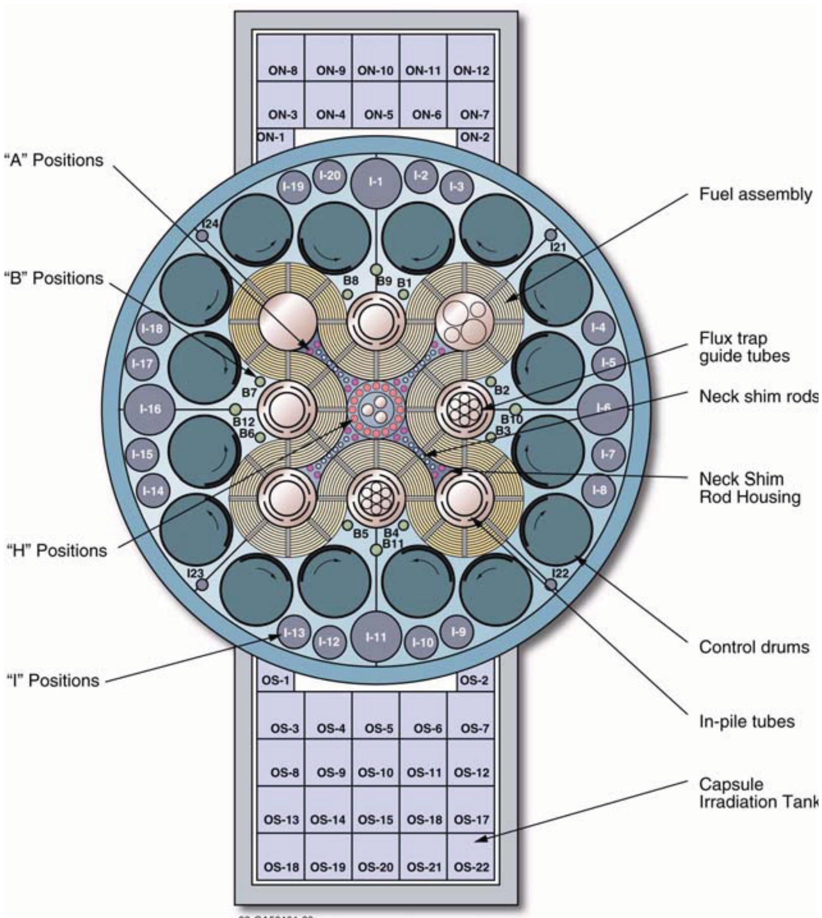


Figure 1. ATR core cross section identifying irradiation locations.

- ***Pressurized Water Loop Experiment -*** Five of the nine ATR flux traps used for materials and fuels testing are equipped with pressurized water loops (at the NW, N, SE, SW, and W locations). A sixth loop will be operational in 2010. Each of the water loops can be operated at different temperatures, pressures, flow rates, or water chemistry requirements. These loops can operate above the standard temperatures and pressure of a commercial PWR power plant. The great advantage of loop tests is the ease with which a variety of samples can be subjected to conditions specified for any PWR design. Each pressurized loop in the ATR is instrumented to measure and control coolant flow, temperatures, pressures and sample test data.

Clearly, the ATR design offers unique advantages for testing. With additional in-pile instrumentation to support these testing capabilities, the features offered by this reactor user facility can be even more fully utilized.

1.2. Approach to Address ATR User Needs for Enhanced Instrumentation

Despite its long history for developing highly specialized instrumentation to meet demands of customers conducting unique tests in one-of-a-kind test facilities, INL instrumentation research funding decreased significantly in the 1980s when large nuclear test facility programs ended. In 2004, an instrumentation effort was restarted that allowed INL to develop unique instrumentation required for ATR irradiations. Because much of the sensor fabrication and evaluation equipment and expertise were still available, INL's High Temperature Test Laboratory (HTTL) staff quickly developed high temperature thermocouples requested by ATR customers for fuel irradiations. Currently, several INL efforts are underway to enhance in-pile instrumentation for ATR users. This section describes the approach being used by INL to identify and prioritize ATR in-pile instrumentation development research.

INL efforts to enhance ATR instrumentation began by first reviewing references (e.g., References [2] through [13]) to identify instrumentation available to users at other test reactors located in the U.S. and abroad. Table I summarizes results from this review. The column labeled "Technology Available at ATR" indicates the types of sensors currently available to ATR users. The column "Proposed Advanced Technology" includes two categories: "Available at Other Reactors" identifies several technologies employed at other test reactors that could be adapted to enhance ATR instrumentation capabilities and "Developmental" identifies developmental or non-nuclear technologies that could be used in irradiation tests. Blue text denotes the instrumentation currently being pursued as part of ATR NSUF research activities, and red text denotes new instrumentation developed by INL and deployed in the ATR. Note that many of these instrumentation development efforts are in collaboration with other organizations. The instrumentation currently being evaluated for the ATR NSUF (denoted by blue text in Table I) was selected based on anticipated user needs and 'technology readiness' (providing ATR users needed instrumentation in the near-term).

Although not discussed in this paper, efforts are also being initiated to develop standardized instrumented lead and PWR test train designs that incorporate new ATR NSUF instrumentation and instrumentation currently used at test reactors. Data from initially deployed standardized test vehicles will be used to validate the performance of developmental instrumentation.

Table I. Review of instrumentation available at ATR and other test reactors

Parameter	Parameter			ATR Technology	Proposed Advanced Technology	
	Static Capsule	Instr. Lead	PWR Loop		Available at Other Reactors	Developmental
Temperature	√	√	√	-Melt wires (peak) -Paint spots (peak)	-SiC Temperature Monitors (range)	-Wireless (range)
		√	√	-Thermocouples (Types N, K, C, and HTIR-TC)[a]		- Fiber Optics
Thermal Conductivity		√	√	-Out-of-pile examinations	-Degradation using signal changes in thermocouples	-Hot wire techniques
Fluence (neutron)	√	√	√	-Flux wires (Fe, Ni, Nb)	-Activating foil dosimeters	
		√	√		-Self-Powered Neutron Detectors (SPNDs) -Subminiature fission chambers	-Moveable SPNDs
Gamma Heating		√	√		-Degradation using signal changes in thermocouples	
Dimensional	√	√	√	-Out-of-pile examinations		
		√	√		-LVDTs (stressed and unstressed) -Diameter gauge -Hyper-frequency resonant cavities	- Ultrasonic Transducers -Fiber Optics
Fission Gas (Amount, Composition)		√	√	-Gas Chromatography -Pressure sensors - Gamma detectors - Sampling	-LVDT-based pressure gauge	-Acoustic measurements with high-frequency echography
Loop Pressure			√	-Differential pressure transmitters -Pressure gauges with impulse lines		
Loop Flowrate			√	-Flow venturis -Orifice plates		
Loop Water Chemistry			√	-Off-line sampling / analysis		
Crud Deposition			√	-Out-of-pile examinations	-Diameter gauge with neutron detectors and thermocouples	
Crack Growth Rate			√		-Direct Current Potential Drop Technique	

[a]Type C thermocouple use requires a "correction factor " to correct for decalibration during irradiation.

2. TEMPERATURE SENSOR DEVELOPMENT EFFORTS

Temperature is a key parameter of interest during fuel and material irradiations. Because of its importance, INL efforts have included several new methods for detecting temperature during irradiation. Using specialized equipment at INL's High Temperature Test Laboratory (HTTL), efforts have focused on implementing two new sensors: (a) unique new thermocouples that resist decalibration due to high temperatures and neutron transmutation in instrumented lead and loop tests; and (b) silicon carbide temperature monitors for static capsule tests. Although not discussed in this paper, INL is also exploring the use of fiber optics as a non-contact temperature sensor.

2.1. High Temperature Irradiation Resistant Thermocouples (HTIR-TCs)

Commercially-available thermocouples drift due to degradation at high temperatures (above 1100 °C) or due to transmutation of thermocouple components. Thermocouples are needed that can withstand both high temperature and high radiation environments. To address this need, INL developed a High Temperature Irradiation Resistant ThermoCouple (HTIR-TC) design that contains commercially-available doped molybdenum paired with a niobium alloy. Battelle Energy Alliance (BEA), the operating contractor for INL, has filed a patent application for this technology, and INL now offers these sensors, with diameters ranging from 1.0 to 3.2 mm, to customers for nuclear and non-nuclear applications. HTIR-TC component materials were selected based on data obtained from materials interaction tests, ductility investigations, and resolution evaluations (see [14] through [17]). To stabilize grain growth, HTIR-TCs are heat treated prior to calibration. Calibration data indicates that HTIR-TCs typically have accuracies of 0.1 to 0.4%. To demonstrate HTIR-TC long duration performance, thermocouples have been heated at elevated temperatures (from 1200 °C to 1800 °C) for up to 6 months. The 1200 °C test included nineteen commercially-available Type N thermocouples, three commercially-available Type K thermocouples, and nine INL-developed swaged HTIR-TCs. As indicated in Figure 2, some Type K and N thermocouples drifted by over 100 °C or 8%. Much smaller drifts (typically less than 20 °C or 2%) were observed in the INL-developed HTIR-TCs. Similar drifts (2%) were observed in HTIR-TCs in a long duration (4000 hour) test completed at 1400 °C.

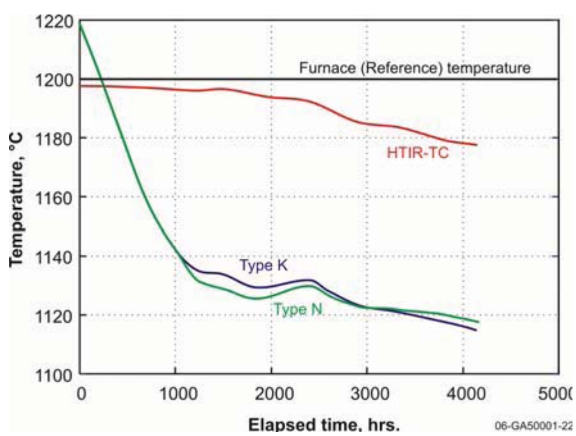


Figure 2. Representative thermocouple response in 1200 °C tests.

HTIR-TCs were installed in a multi-capsule experiment that is currently being irradiated in INL's ATR. This multi-capsule experiment is designed to irradiate samples at temperatures up to approximately 1200 °C. This test, which started in February 2007, is still underway. Figure 3 shows signals from one of the test capsules containing two INL-developed HTIR-TCs and one Type N thermocouple. The Type N thermocouple was purposely located at a cooler region within this test capsule. Signal variations are due to ATR power fluctuations and outages. At the beginning of this irradiation, the HTIR-TC located near the Type N thermocouple gave a signal consistent with the signal from the Type N thermocouple. In addition, the HTIR-TC located at a hotter region within the capsule is yielding a consistent signal, but at a higher temperature. However, in October 2008, the Type N thermocouple failed and its signal ceased.

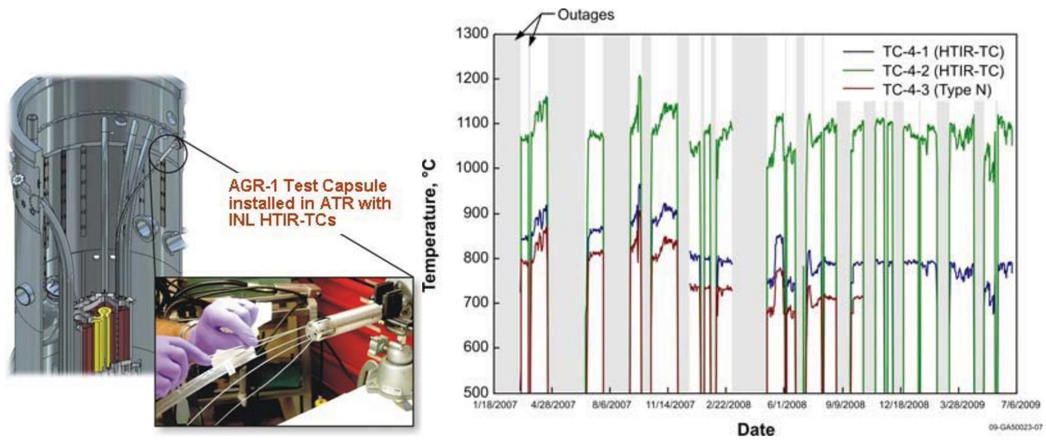


Figure 3. HTIR-TCs installed in AGR-1 test capsule and representative HTIR-TC and Type N data during ATR irradiation.

Since its initial development, INL has continued to investigate options that could reduce fabrication costs and enhance the reliability of INL-developed HTIR-TCs for higher temperature in-pile testing, allowing their use in applications up to 1800 °C. Investigations identified several improvements in HTIR-TC fabrication processes that not only reduced costs but also improved thermocouple reliability, such as automated, consistent fabrication techniques, alternate components in compensating soft extension cable, and improved heat treatments. Detailed information about earlier options explored in these efforts is documented in References [15] through [17]. More recent results from efforts to investigate alternate designs of HTIR-TCs that offer the potential to improve HTIR-TC operation at higher temperatures are reported in this paper.

Initially-deployed HTIR-TCs rely on swaging fabrication techniques because of their simplicity and durability. As shown in Figure 4a, a swaged TC is fabricated by loading pre-formed, crushable insulator beads onto thermoelement wires and placing the insulated thermoelements in a sheath (tube) that is then swaged (compacted) to form a single, cohesive component. If desired, the thermocouple may be joined to hard extension cable using a splice sleeve as shown in Figure 4a. Recently, INL has explored drawn and loose assembly HTIR-TC designs. Drawn thermocouples are prepared similarly to swaged thermocouples in that crushable insulator beads are loaded onto thermoelement wires that are placed in a sheath. However, the drawn assembly is passed through a stationary die to form a thermocouple. In a loose assembly thermocouple (see Figure 4b), preformed hard-fired insulators are loaded onto thermoelement wires. After being placed in a sheath, a splice sleeve is used to join the thermocouple assembly to the hard extension cable. To preclude any oxidation of thermocouple components, the assembly is vacuum purged and backfilled with inert gas through this splice sleeve.

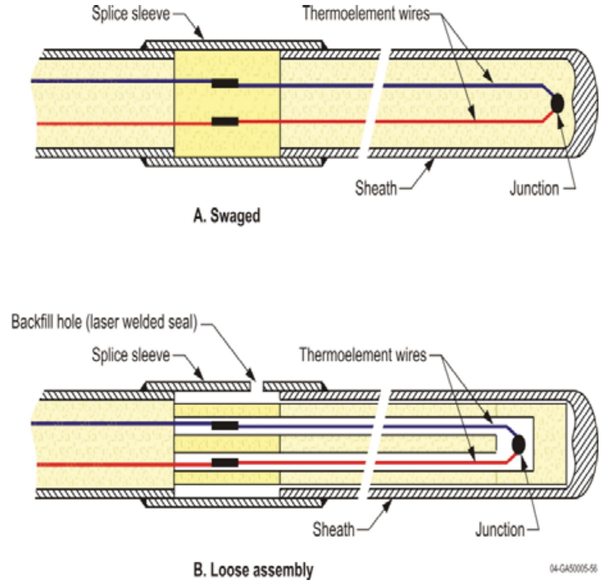


Figure 4. Swaged and loose assembly thermocouple configurations.

Drawn and loose assembly thermocouple designs each offer the potential for improved high temperature performance over swaged thermocouples. Compared to swaging, the drawing process does not twist HTIR-TC thermoelement wires during fabrication. In addition, INL evaluations indicate that drawing leads to less distortion of thermoelement wires. In a loose assembly HTIR-TC, higher temperatures may be possible because thermoelement thinning and irregular deformation associated with swaging and drawing are avoided. Furthermore, the loose fit can accommodate some differential thermal expansion of thermocouple components without inducing thermoelement stress.

Drawn and loose assembly HTIR-TC fabrication requires specialized fixturing and components. INL designed and installed a specialized draw bench at the HTTL for drawn thermocouple fabrication (see Figure 5). Loose assembly HTIR-TC fabrication requirements included developing specialized splice sleeve and insulators for joining the loose assembly thermocouple to the hard extension cable. As shown in Figure 6, three sealing welds are also needed (connecting the two halves of the splice sleeve, between the extension cable sheath and the splice sleeve, and between the splice sleeve and the TC sheath). At this point, a vacuum/backfill process is used to fill the thermocouple sheath with an inert gas using the specialized fixturing shown in Figure 6. In this specialized fixturing, the assembly is vacuumed down to ~10^{-5} torr and backfilled with helium through a tiny hole in the splice sleeve. The backfill hole in the splice sleeve is welded closed through the view port of the fixturing using a laser welder.

Figure 5. HTTL fixturing for fabricating drawn HTIR-TCs.

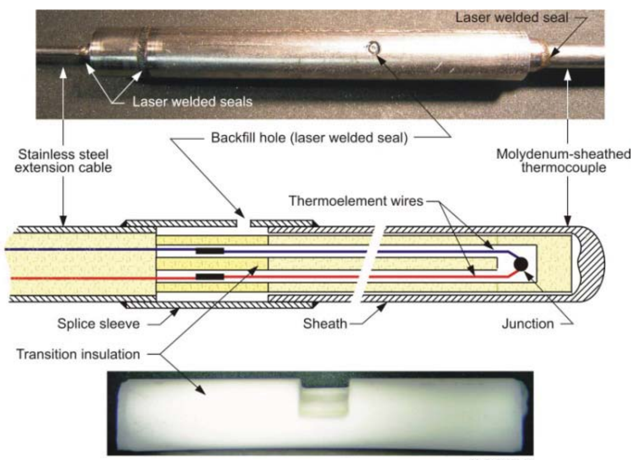
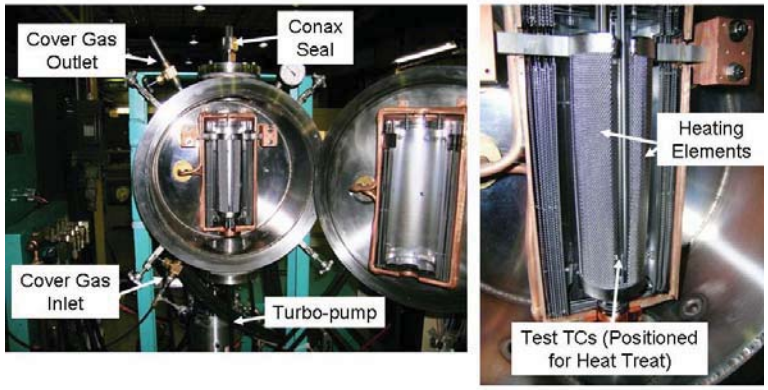


Figure 6. Specialized loose assembly HTIR-TC components.

A long duration (1000 hour) test was recently completed to compare the performance of swaged, drawn, and loose assembly HTIR-TCs at 1500 °C. This test was performed in a high temperature vacuum furnace, pictured in Figure 7. Multiple swaged, drawn, and loose assembly thermocouples were included in this test.



(a) Vacuum Chamber (b) Test TC Installation

Figure 7. Vacuum furnace test setup.

Results from the 1500 °C test are shown in Figure 8. The signals from the swaged and drawn HTIR-TCs decreased by 2.3%, primarily during the first 600 hours of the test. The loose assembly thermocouple displayed greater stability and resolution than either the swaged or drawn thermocouples. Although the swaged HTIR-TC response is consistent with the decalibration observed in prior 1200 and 1400 °C 4000 hour tests, the fact that most of the drift occurred during initial portions of this test suggests that longer duration heat treatments may be needed to stabilize the distortion associated with swaging or drawing processes in larger diameter thermoelement wires (prior HTTL efforts to optimize heat treatment processes had focused on thermocouples smaller diameter wires).

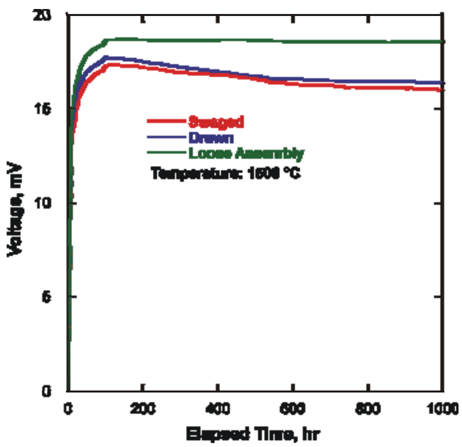


Figure 8. Thermocouple performance after 1000 hours at 1500 °C

Selected thermocouples from this test have also been tested at 1800 °C without any additional heat treatment. Results, shown in Figure 9, suggest that HTIR-TCs are also capable of functioning at these temperatures. Although a furnace malfunction led to this test being terminated prematurely at 170 hours, the loose assembly and drawn HTIR-TCs exhibited less than 1 % drift and the swaged HTIR-TC exhibited approximately 8% drift. Note that fabrication processes for HTIR-TCs expected to operate at 1800 °C would include heat treatment above this temperature. Although better stability would be expected from all three HTIR-TC designs if they had been heat treated for operation at 1800 °C, the performance of all three HTIR-TC designs in this test suggests that they are viable for this temperature.

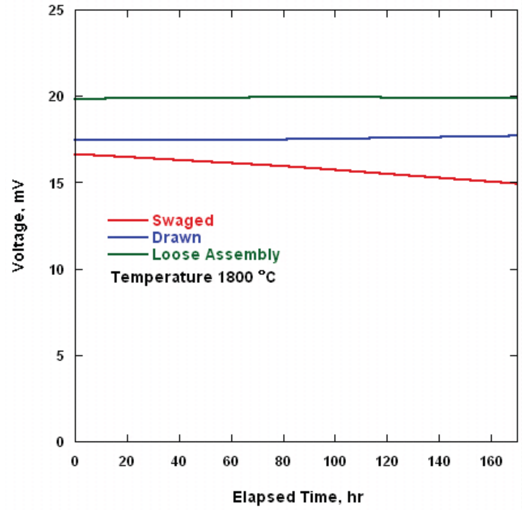


Figure 9. Thermocouple performance at 1800 °C

2.2. Silicon Carbide Temperature Monitors

As noted in Table I, silicon carbide (SiC) temperature monitors are being explored by INL for use as temperature sensors in ATR static capsules. Melt wires or paint spots, which are typically for temperature sensors in static capsules, are limited because they can only detect whether a single temperature was exceeded or not. SiC temperature monitors are advantageous because a single monitor can be used to detect for a range of temperatures that may have occurred during irradiation.

For decades, post-irradiation temperature monitors have been based on the phenomenon that irradiation-induced defects of SiC begin to anneal out at temperatures exceeding its irradiation temperature. These SiC monitors have relied on changes in length, density, thermal conductivity, and electrical resistivity to infer irradiation temperature. However, Snead et al. [13] recommends using changes in resistivity because of improved accuracy, ease of measurement, and reduced costs. Experimental data indicate that accuracies of approximately 20 °C are possible with this technique for dose ranges of 1 to 8 dpa and for temperatures of at least between 200 and 800 °C. Experimental data reported in [13] suggest that upper and lower bounds for this range may be extended.

A capability similar to the technique reported in [13] is being implemented at INL. Specialized equipment at INL's HTTL has been configured so that it can be used to measure resistivity of SiC samples after heating at various temperatures. SiC electrical resistivity is measured after heating in a furnace located within a stainless steel enclosure at the HTTL (see Figure 10). SiC monitors are heated in a furnace that is placed under a hood located within the stainless steel enclosure. Temperature accuracy in this furnace is verified using thermocouples that have been calibrated using National Institute of Standards and Testing (NIST)-traceable sources. The ventilation system is activated during heating so that any vapors that offgas during heating are vented through this ventilation system. After heating, cooled samples are placed into a constant temperature environmental test chamber to insure electrical resistivity measurements are taken within 0.2 °C of a predetermined temperature, 30 °C. Reference [13] indicates that resistance measurements must be taken at nearly the same temperature or it will adversely affect the accuracy of SiC temperature monitors. A high accuracy (9 digits) multimeter is used to obtain resistance measurements. This multimeter is placed outside the stainless steel enclosure.

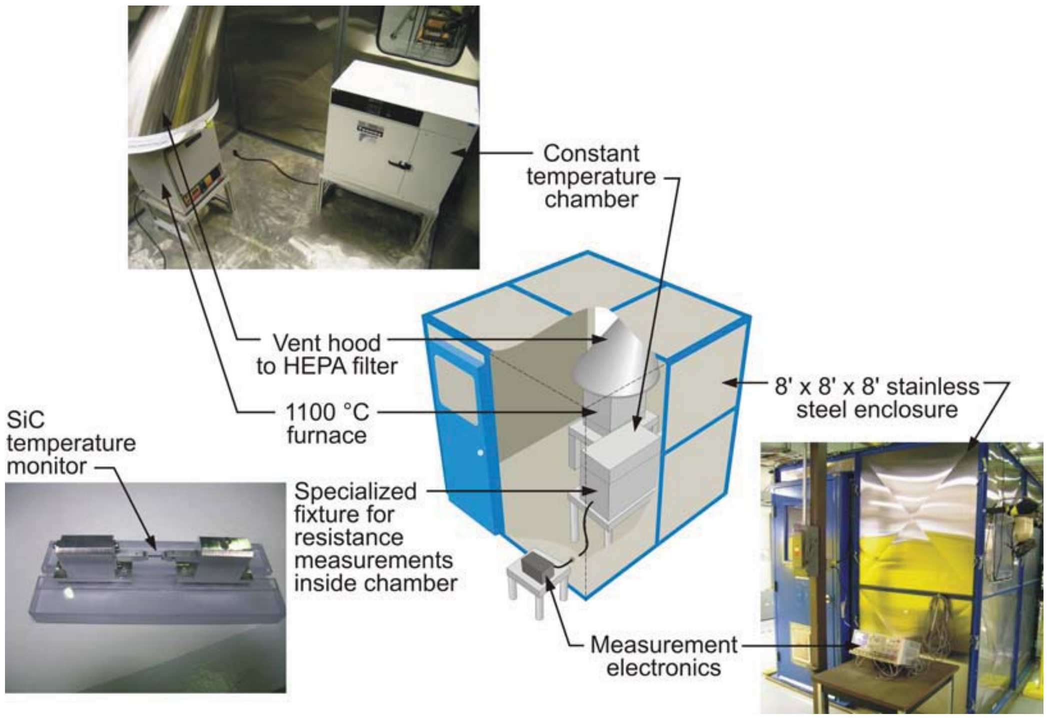


Figure 10. Setup for annealing and measuring electrical resistivity of SiC temperature monitors.

Reference 2 indicates that resistance measurements should be taken with SiC temperature monitors always placed with the same orientation. In addition to marking on the SiC monitors, special fixturing has been developed to facilitate these measurements. A four point probe technique is used with the four points contacted to the sample through spring-loaded angled electrodes that hold the SiC carbide temperature monitor in place, as shown in Figure 10. Current and voltage are provided to the sample through wires that are threaded through the holes in the electrodes.

Temperature monitors, approximately 1 mm x 1 mm x 10 mm, for ATR irradiations were fabricated from high grade SiC (SC-003) manufactured by Rohm and Haas. [1] ATR capsules containing SiC temperature monitors will be used to irradiate materials of interest for advanced reactor applications at a variety of temperatures (nominally 300 °C, 400 °C, 500 °C, and 700 °C) and total dose accumulations (nominally 3 dpa and 6 dpa). In September 2008, such a capsule was inserted in the ATR East Flux Trap, where the fast flux is approximately 9.7×10^{13} n/cm^2-s (E > 1 MeV). Although a variety of materials are included in this capsule, a dose of 1 dpa in stainless steel roughly corresponds to a fluence of 7×10^{20} n/cm^2.

[1] References herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the U.S. Government, any agency thereof, or any company affiliated with the Idaho National Laboratory.

To verify the operability of equipment installed at the HTTL, INL tested several samples made from two grades of SiC produced by Rohm and Haas: SC-002, which has an average resistivity of 1 ohm-cm; and SC-003 which has an average resistivity of 8323 ohm-cm (actual vendor measurements for SC-003 varied between 2564 to 23,886 ohm-cm). Results indicate that INL can successfully detect which grade of SiC is in a temperature monitor. In addition, INL was able to obtain consistent resistivity data for samples made from each grade of SiC (e.g., resistivity values were within 13% of the average measured value for each grade of SiC). Furthermore, the measured values for SC-003 were within the range of values provided by the vendor for this run of material. To further verify the accuracy of the INL setup, a joint INL/ORNL effort has been initiated to perform comparison runs of irradiated SiC temperature monitors.

3. CONCLUSIONS

An effort is underway to provide enhanced in-pile instrumentation for ATR users. This effort to enhance ATR instrumentation began by first completing a review to identify what instrumentation was available at other test reactors in the world. Developmental or non-nuclear technologies that could be used in ATR irradiation tests were also considered. Instrumentation development activities were then prioritized based on anticipating customer needs and technology readiness. In addition, instrumentation development collaborations were begun with other organizations that employ similar sensors in their test facilities. Efforts are also underway to standardize instrumented lead and PWR test train designs that incorporate this instrumentation. Data from initially deployed standardized test vehicles will be used to validate the performance of developmental instrumentation.

This effort has resulted in several new sensors now being available to ATR NSUF users and other research organizations. Representative results from on-going INL efforts to evaluate sensors for detecting temperature during irradiation testing that are presented in this paper illustrate the process used within this project.

ACKNOWLEDGMENTS

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